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TRANSMITTAL LETTER TO THE UNITED STATES

ATTORNEY'S DOCKET NUMBER 49633

DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

INTERNATIONAL APPLICATION NO.  
PCT/EP 99/09971INTERNATIONAL FILING DATE  
15 December 1999PRIORITY DATE CLAIMED  
15 December 1998

TITLE OF INVENTION: REACTOR MODULE HAVING A CATALYST-TUBE BUNDLE

APPLICANT(S) FOR DO/EO/US Gerhard OLBERT, Franz CORR

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. /X/ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
  2. / / This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
  3. /X/ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
  4. /X/ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
  5. /X/ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
    - a./X/ is transmitted herewith (required only if not transmitted by the International Bureau).
    - b./ / have been transmitted by the International Bureau.
    - c./ / is not required, as the application was filed in the United States Receiving Office (RO/USO).
  6. /X/ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
  7. /X/ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
    - a./X/ are transmitted herewith (required only if not transmitted by the International Bureau).
    - b./ / have been transmitted by the International Bureau.
    - c./ / have not been made; however, the time limit for making such amendments has NOT expired.
    - d./ / have not been made and will not be made.
  8. /X/ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
  9. /X/ An oath or declaration of the inventor(s) (35 U.S.C. 171(c)(4)).
  10. / / A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
- Items 11. to 16. below concern other document(s) or information included:
11. / / An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
  12. /X/ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
  13. /X/ A FIRST preliminary amendment.  
/ / A SECOND or SUBSEQUENT preliminary amendment.
  14. / / A substitute specification.
  15. / / A change of power of attorney and/or address letter.
  16. /X/ Other items or information.  
International Search Report  
International Preliminary Examination Report

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U.S. Appl. No. (If Known) INTERNATIONAL APPLN. NO.  
PCT/EP99/09971ATTORNEY'S DOCKET NO.  
49633

		CALCULATIONS	PTO USE ONLY
17. /X/ The following fees are submitted			
BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):			
Search Report has been prepared by the EPO or JPO.....	\$860.00	860.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482).....	\$750.00		
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....	\$700.00		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO .....	\$ 970.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4).....	\$96.00		
ENTER APPROPRIATE BASIC FEE AMOUNT = \$		860.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than / / 20 / / 30 months from the earliest claimed priority date (37 CFR 1.492(e)).			
Claims	Number Filed	Number Extra	Rate
Total Claims	10 -20		X\$18.
Indep. Claims	1 -3		X\$80.
Multiple dependent claim(s) (if applicable)		+270.	
TOTAL OF ABOVE CALCULATION		=	860.00
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).			
SUBTOTAL		=	860.00
Processing fee of \$130. for furnishing the English translation later than / / 20 / / 30 months from the earliest claimed priority date (37 CFR 1.492(f)).			
TOTAL NATIONAL FEE		=	860.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) \$40.00 per property			
TOTAL FEES ENCLOSED		= \$	900.00
Amount to be refunded:		\$	
Charged		\$	

a./X/ A check in the amount of \$ 900. to cover the above fees is enclosed.

b./ / Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \$ \_\_\_\_\_ to cover the above fees. A duplicate copy of this sheet is enclosed.

c./X/ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 11-0345. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:  
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SIGNATURE

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NAME

Registration No. 18,967

09/868093

JC03 Rec'd PCT/TL 14 JUN 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of  
OLBERT et al.

BOX PCT

International Application  
PCT/EP 99/09972

Filed: December 15, 1999

For: REACTOR MODULE HAVING A CATALYST-TUBE BUNDLE

PRELIMINARY AMENDMENT

Honorable Commissioner of  
Patents and Trademarks  
Washington, D.C. 20231

Sir:

Prior to examination, kindly amend the above-identified application as follows:

IN THE CLAIMS

Kindly amend the claims as shown in the attached sheet.


REMARKS

The claims were amended in the preliminary examination. The claims have been amended further to eliminate multiple dependency and to put them in better form for U.S. filing. No new matter is included. A clean copy of the claims is attached.

Favorable action is solicited.

Respectfully submitted,

KEIL & WEINKAUF

  
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(202)659-0100

CLEAN COPY OF AMENDED CLAIMS - OZ49633

3. A reactor module (1) as claimed in claim 1, which has a length:width ratio of from 1:1 to 10:1, preferably from 3:1 to 6:1, particularly preferably 5:1.
4. A reactor module ( 1) as claimed in claim 1, which has an odd number of baffle plates (9), preferably 1, 3 or 5 baffle plates (9).
6. A reactor module as claimed in claim 2, which has one or more bypasses with fixed (11) or adjustable (10) passage apertures, through the baffle plates (9), in their tube-free regions.
7. A reactor module (1) as claimed in claim 1, wherein intermediate walls (12), which form in each case a lower, outer prechamber (13) and a lower, inner prechamber (14) in the feed line (3) and an upper, outer prechamber (15) and an upper, inner prechamber (16) in the discharge line (4), are provided in the feed and discharge lines (3,4), and wherein the heat-exchange medium is fed to the lower, outer prechamber (13), through a region between the feed line (3) and the discharge line (4) to the upper, inner prechamber (16), through its jacket aperture (6) to the reactor space surrounding the catalyst tubes (2), subsequently through the jacket aperture (5) to the lower, inner prechamber (14), through the region between the feed and discharge lines (3, 4) to the upper, outer prechamber (15), and finally back to the pump(s) through the discharge line (4).
9. A reactor module (1) as claimed in claim 7, wherein the catalyst tubes of the catalyst-tube bundle (2) are arranged in mutually offset rows, where the ratio of the tube separation  $S_t$  transverse to the inflow direction by the heat-exchange

AMENDED CLAIMS - MARKED UP VERSION OZ49633

medium to the tube separation  $S_1$  longitudinal to the inflow direction by the heat-exchange medium is preferably greater than or equal to  $2\sqrt{3}$ , particularly preferably equal to  $2\sqrt{3}$ .

10. A reactor constructed from two or more reactor modules (1) as claimed in claim 7 lined up in rows against one another on the narrow side surfaces in the direction of the longitudinal axes of the catalyst tubes.
11. The use of a reactor module as claimed in claim 1 [one of claims 1 to 9] or of a reactor as claimed in claim 10 for carrying out oxidation reactions, in particular for the preparation of phthalic anhydride, maleic anhydride, glyoxal, (meth)acrolein or (meth)acrylic acid.

## AMENDED CLAIMS - MARKED UP VERSION

3. A reactor module (1) as claimed in claim 1 [one of claims 1 and 2], which has a length:width ratio of from 1:1 to 10:1, preferably from 3:1 to 6:1, particularly preferably 5:1.
4. A reactor module (1) as claimed in claim 1 [one of claims 2 and 3], which has an odd number of baffle plates (9), preferably 1, 3 or 5 baffle plates (9).
6. A reactor module as claimed in claim 2 [one of claims 2 to 5], which has one or more bypasses with fixed (11) or adjustable (10) passage apertures, through the baffle plates (9), in their tube-free regions.
7. A reactor module (1) as claimed in claim 1 [one of claims to 6], wherein intermediate walls (12), which form in each case a lower, outer prechamber (13) and a lower, inner prechamber (14) in the feed line (3) and an upper, outer prechamber (15) and an upper, inner prechamber (16) in the discharge line (4), are provided in the feed and discharge lines (3,4), and wherein the heat-exchange medium is fed to the lower, outer prechamber (13), through a region between the feed line (3) and the discharge line (4) to the upper, inner prechamber (16), through its jacket aperture (6) to the reactor space surrounding the catalyst tubes (2), subsequently through the jacket aperture (5) to the lower, inner prechamber (14), through the region between the feed and discharge lines (3, 4) to the upper, outer prechamber (15), and finally back to the pump(s) through the discharge line (4).
9. A reactor module (1) as claimed in claim 7 [one of claims 1 to 8], wherein the catalyst tubes of the catalyst-tube bundle (2) are arranged in mutually offset

AMENDED CLAIMS - MARKED UP VERSION OZ49633

rows, where the ratio of the tube separation  $S_t$  transverse to the inflow direction by the heat-exchange medium to the tube separation  $S_l$  longitudinal to the inflow direction by the heat-exchange medium is preferably greater than or equal to  $2\sqrt{3}$ , particularly preferably equal to  $2\sqrt{3}$ .

10. A reactor constructed from two or more reactor modules (1) as claimed in claim 7 [one of claims 7 to 9] lined up in rows against one another on the narrow side surfaces in the direction of the longitudinal axes of the catalyst tubes.
11. The use of a reactor module as claimed in claim 1 [one of claims 1 to 9] or of a reactor as claimed in claim 10 for carrying out oxidation reactions, in particular for the preparation of phthalic anhydride, maleic anhydride, glyoxal, (meth)acrolein or (meth)acrylic acid.

CURRENT CLAIMS - OZ 49633

1. A reactor module (1) having a rectangular cross section having a catalyst-tube bundle (2) of from 10,000 to 40,000 catalyst tubes through whose space surrounding the catalyst tubes a heat-exchange medium circuit is run, with feed and discharge lines (3,4) at both ends of the reactor module with jacket apertures (5, 6) for the feed and discharge respectively of a heat-exchange medium in cross-current to the catalyst tubes by means of one or more pumps (P), if desired while passing the heat-exchange medium or a sub-stream of the heat-exchange medium through one or more external heat exchangers (W), where the heat-exchange medium is fed to the lower line (3) and is fed back to the pump(s) (P) via the upper line (4), and having catalyst tube-free spaces (7, 8), which preferably extend over the entire reactor height, arranged in the reactor space on two opposite reactor side surfaces which are parallel to the catalyst tubes, with one or more baffle plates (9), which leave passage cross sections open alternately in the spaces (7, 8).
2. A reactor module as claimed in claim 1, wherein the catalyst tube-free spaces (7, 8) are arranged on the two broad reactor side surfaces.
3. A reactor module (1) as claimed in claim 1, which has a length:width ratio of from 1:1 to 10:1, preferably from 3:1 to 6:1, particularly preferably 5:1.
4. A reactor module (1) as claimed in claim 2, which has an odd number of baffle plates (9), preferably 1, 3 or 5 baffle plates (9).
5. A reactor module (1) as claimed in claim 4, wherein the pump(s) and, if desired, the external heat exchanger(s) are arranged on the same, preferably broad, side





10. A reactor constructed from two or more reactor modules (1) as claimed in claim 7 lined up in rows against one another on the narrow side surfaces in the direction of the longitudinal axes of the catalyst tubes.
11. The use of a reactor module as claimed in claim 1 or of a reactor as claimed in claim 10 for carrying out oxidation reactions, in particular for the preparation of phthalic anhydride, maleic anhydride, glyoxal, (meth)- acrolein or (meth)acrylic acid.

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## AS ORIGINALLY FILED

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### Reactor module having a catalyst-tube bundle

- 10 The present invention relates to a reactor module having a catalyst-tube bundle, to a reactor constructed from two or more reactor modules lined up in rows against one another, and to the use of a reactor module or a reactor for carrying out oxidation reactions.
- 15 The usual design of reactors of the generic type consists of a generally cylindrical tank, in which a bundle, i.e. a multiplicity, of catalyst tubes is accommodated, usually in a vertical arrangement. These catalyst tubes, which may, if desired, contain supported catalysts, are attached in a sealing manner with their ends in tube plates and in each case open at the upper or lower end into a hood connected to the
- 20 tank. The reaction mixture flowing through the catalyst tubes is fed in and out via these hoods. A heat-exchange medium circuit runs through the space surrounding the catalyst tubes in order to equalize the heat balance, in particular in the case of highly exothermic reactions.
- 25 For economic reasons, reactors having a very large number of catalyst tubes are employed, the number of catalyst tubes accommodated frequently being in the range from 10,000 to 40,000 (cf. DE-A-44 31 949).
- 30 The technical upper limit of the reactor diameter and thus the number of tubes is thus reached with respect to manufacture, transport, installation and reaction engineering, in particular equal distribution of the coolant. Since the generally gaseous reaction mixture is usually under pressure, semicircular shapes of the hoods delimiting the gas space have proven successful at the lower and in particular at the upper end of the tank. For these hood shapes, a cylindrical
- 35 structure of the reactor is a prerequisite.

The cylindrical reactor geometry is associated with the disadvantage that, in particular in the case of the technologically particularly advantageous cross flow of the heat-exchange medium to the catalyst tubes from a region outside the catalyst tubes to the catalyst tube-free interior of the reactor, the cross-sectional area, which  
5 decreases greatly in the radial direction toward the inside, does not allow the full coolant stream to run into the interior of the catalyst-tube bundle. Instead, coolant must be discharged via holes in the baffle plates in order to keep the pressure loss and thus the pump performance within acceptable limits.

10 It is an object of the present invention to facilitate a constant heat-exchange medium flow over the reactor cross section.

In one embodiment, it is an object of the present invention to provide a reactor whose capacity can be matched to the requirements of the individual case.

15 We have found that this object is achieved by a reactor module having a catalyst-tube bundle through whose space surrounding the catalyst tubes a heat-exchange medium circuit is run, with feed and discharge lines at both ends of the reactor module with jacket apertures for the feed and discharge respectively of a heat-  
20 exchange medium in cross-current to the catalyst tubes by means of one or more pumps, if desired while passing the heat-exchange medium or a sub-stream of the heat-exchange medium through one or more external heat exchangers, where the heat-exchange medium is fed to the lower line and is fed back to the pump(s) via the upper line, wherein the reactor module has a rectangular cross section.

25 It has been found that hoods delimiting a pressurized gas space can also be designed with a semicylindrical geometry, as necessary for both-sided delimitation of a tank with a rectangular cross section.

30 In a preferred manner, catalyst tube-free spaces, which preferably extend over the entire reactor height, are arranged in the reactor space on two opposite reactor side surfaces which are parallel to the catalyst tubes, and one or more baffle plates, which leave passage cross sections open alternately in the catalyst tube-free spaces. Through this design, the heat-exchange medium stream can be fed uniformly, in  
35 the desired manner, around the catalyst tubes.

The catalyst tube-free spaces are preferably arranged at the two broad reactor side surfaces.

5 With respect to the relative dimensions of the length and width of the reactor module, a ratio of from 1:1 to 10:1 is advantageous, preferably from 3:1 to 6:1, particularly preferably 5:1. For the reactor height, relative values of from 1.5 m to 7 m are preferred.

10 An odd number of baffle plates is preferably provided; this causes the feed and discharge of the heat-exchange medium to take place on the same side of the reactor module. 1, 3 or 5 baffle plates are particularly preferably provided.

15 It is advantageous to arrange the pump(s) and any external heat exchanger(s) on the same, preferably broad, side of the reactor module. This results in a particularly space-saving arrangement.

20 The respective course of the reaction may require different heat profiles; matching to the requirements of the individual case is possible by adjusting the heat-exchange medium stream by means of one or more bypasses through the baffle plates in their tube-free regions, with fixed or adjustable passage apertures.

25 A particularly preferred embodiment is a reactor module with intermediate walls in the feed and discharge lines, which in each case form a lower, outer prechamber and a lower, inner prechamber in the feed line and an upper, outer prechamber and an upper, inner prechamber in the discharge line. The heat-exchange medium is fed to the lower, outer prechamber, through a region between the feed line and discharge line to the upper, inner prechamber, through the jacket aperture thereof to the reactor space surrounding the catalyst tubes, subsequently through a jacket aperture to the lower, inner prechamber, through the region between the feed line and discharge line to the upper, outer prechamber, and finally back to the pump(s) through the discharge line. This facilitates the particularly favorable cocurrent transport of heat-exchange medium and reaction mixture without changing the usual pump arrangement.

35 Matching to the temperature profile required in each case can be achieved in the case of reactors with cocurrent procedure by means of an outside chamber against the broad reactor side assigned to the catalyst tube-free space, with apertures to the

reactor space surrounding the catalyst-tube bundle, and with fixed or adjustable passage apertures for the heat-exchange medium in the outside chamber.

5 A particularly advantageous embodiment proposes a tube division according to which the catalyst tubes of the catalyst-tube bundle are arranged in mutually offset rows, where the ratio of the tube separation  $s_t$  transverse to the inflow direction by the heat-exchange medium to the tube separation  $s_l$  longitudinal to the inflow direction by the heat-exchange medium is preferably greater than or equal to  $2 \cdot \sqrt{3}$ , particularly preferably equal to  $2 \cdot \sqrt{3}$ . A tube arrangement of this type creates  
10 less resistance to the inflowing heat-exchange medium; the pressure loss is correspondingly less at the same time as a higher heat transfer coefficient.

The invention also relates to a reactor constructed from two or more reactor modules lined up in rows against one another on the narrow side surfaces in the  
15 direction of the longitudinal axes of the catalyst tubes. Reactors of this type are distinguished by a flexible capacity, which can be adjusted to the specific requirements. By connecting the reactor modules in series on the narrow side surfaces, the semicylindrical hoods delimiting the gas space can be extended on one of their planar side surfaces, which are correspondingly provided with passage  
20 apertures. An upper limit for the capacity of reactors is thus increased.

The heat-exchange medium circuit can serve equally for dissipation and supply of heat from or to the reaction mixture flowing through the catalyst tubes; the reactor module according to the invention or the reactor according to the invention can  
25 thus be employed for exothermic and for endothermic reactions. They are particularly suitable for carrying out oxidation reactions, in particular for the preparation of phthalic anhydride, maleic anhydride, glyoxal, (meth)acrolein or (meth)acrylic acid.

30 The invention is explained in greater detail below with reference to working examples and a drawing, in which:

Figure 1 shows a diagrammatic representation of a reactor model according to the invention,

35

Figure 2 shows a longitudinal section through a reactor module according to the invention,

- Figure 3 shows a longitudinal section through a preferred embodiment of a reactor module according to the invention,
- 5 Figure 4 shows a longitudinal section through a further preferred embodiment of a reactor module according to the invention,
- Figure 5 shows a preferred arrangement of the catalyst tubes, and
- 10 Figure 6 shows a reactor constructed by way of example from three reactor modules.

Figure 1 shows a reactor module 1 with rectangular cross section having a vertical catalyst-tube bundle 2, with feed line 3 and discharge line 4 for the heat-exchange medium, and with jacket apertures 5, 6 to the reactor module 1. Catalyst tube-free spaces 7, 8 are provided on the opposite broad side surfaces of the reactor module for distribution or collection of the heat-exchange medium. The baffle plates 9 produce a meander-shaped route of the heat-exchange medium. The gas or gas mixture G is fed into the gas inlet space 21, flows through the catalyst tubes 2 and is subsequently discharged via the gas outlet collector 22. Pumps P and heat exchangers W are arranged on the same broad side of the reactor module 1.

In the particular embodiment shown in longitudinal section in Figure 2, bypasses for the heat-exchange medium stream are additionally shown in the baffle plates 9, in their catalyst tube-free regions. These bypasses leave adjustable passage apertures 10 or fixed passage apertures 11 open for the heat-exchange medium.

Figure 3 shows a longitudinal section through a preferred embodiment, with cocurrent transport of heat-exchange medium and gas mixture G. To this end, in each case a lower, outer prechamber 13, a lower, inner prechamber 14 and an upper, outer prechamber 15 and an upper, inner prechamber 16 are formed by means of intermediate walls 12 in the feed and discharge lines 3, 4. The heat-exchange medium is then fed from the feed line 3 into the lower, outer prechamber 13, through the region between the feed and discharge lines 3, 4 to the upper, inner prechamber 16, through the jacket aperture 5 to the space surrounding the catalyst tubes, and subsequently discharged to the pump(s) through the jacket aperture 6, the lower, inner prechamber 14, a region between the feed and discharge lines 3, 4

and the upper, outer prechamber 15. An outer chamber 17 with apertures 18, 19 to the reactor module or to the catalyst tube-free space 8 can preferably be arranged on the broad reactor outside opposite the divided prechambers 13 and 16. Part of the heat-exchange medium stream can be short-circuited via the outer chamber 17, where the heat-exchange medium stream can be adjusted via fixed or adjustable passage apertures 20.

In the particular embodiment shown in longitudinal section in Figure 4, one or more heat exchangers W, via which the sub-streams of the heat-exchange medium are passed from the catalyst tube-free space 8, are arranged on the broad side of the reactor module 1 opposite the pump(s).

Figure 5 shows a cross section through a reactor module with a particularly favorable tube arrangement. In this arrangement, the tubes are arranged in mutually offset rows, with the ratio between the tube separation  $s_t$  transverse to the inflow direction by the heat-exchange medium and the tube separation  $s_l$  longitudinal to the inflow direction by the heat-exchange medium being in the ratio  $2 \cdot \sqrt{3}$ . The tube separation  $s_d$  diagonally to the inflow direction by the heat-exchange medium is then correspondingly less than the tube separation  $s_t$ .

Figure 6 shows by way of example a reactor constructed by connecting three reactor modules 1 in series. In a space-saving manner, all pumps P and heat exchangers W are arranged on the same side of the reactor modules.

The invention ensures a constant heat-exchange medium stream over the reactor cross section. A uniform heat transfer coefficient to the reaction mixture flowing through the catalyst tubes, and thus advantageous performance of the reaction, is thereby achieved.

The design according to the invention reduces the pressure loss by up to half compared with conventional designs. The economic efficiency is thus improved, since lower pump capacities or higher heat-exchange medium circulation quantities are possible.

A further reduction in the pressure loss is achieved by the particularly favorable, offset tube distribution, with the narrowest cross sections in the diagonals to the flow direction by the heat-exchange medium.



A further advantage of the apparatus according to the invention is its modular design, i.e. reactors with any desired capacity can be provided by connecting a corresponding number of reactor modules in series.

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	100
1	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	100
2	2	1	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	100
3	3	2	1	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	100
4	4	3	2	1	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	100
5	5	4	3	2	1	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80																				

**AS ENCLOSED TO IPER**

We claim:

- 5 1. A reactor module (1) having a rectangular cross section having a catalyst-tube bundle (2) of from 10,000 to 40,000 catalyst tubes through whose space surrounding the catalyst tubes a heat-exchange medium circuit is run, with feed and discharge lines (3, 4) at both ends of the reactor module with jacket apertures (5, 6) for the feed and discharge respectively of a heat-exchange medium in cross-current to the catalyst tubes by means of one or more pumps (P), if desired while passing the heat-exchange medium or a sub-stream of the heat-exchange medium through one or more external heat exchangers (W), where the heat-exchange medium is fed to the lower line (3) and is fed back to the pump(s) (P) via the upper line (4), and having catalyst tube-free spaces (7, 8), which preferably extend over the entire reactor height, arranged in the reactor space on two opposite reactor side surfaces which are parallel to the catalyst tubes, with one or more baffle plates (9), which leave passage cross sections open alternately in the spaces (7, 8).
- 10
- 15
- 20 2. A reactor module as claimed in claim 1, wherein the catalyst tube-free spaces (7, 8) are arranged on the two broad reactor side surfaces.
3. A reactor module (1) as claimed in one of claims 1 and 2, which has a length:width ratio of from 1:1 to 10:1, preferably from 3:1 to 6:1, particularly preferably 5:1.
- 25
4. A reactor module (1) as claimed in one of claims 2 and 3, which has an odd number of baffle plates (9), preferably 1, 3 or 5 baffle plates (9).
- 30 5. A reactor module (1) as claimed in claim 4, wherein the pump(s) and, if desired, the external heat exchanger(s) are arranged on the same, preferably broad, side of the reactor module (1).
- 35 6. A reactor module as claimed in one of claims 2 to 5, which has one or more bypasses with fixed (11) or adjustable (10) passage apertures, through the baffle plates (9), in their tube-free regions.

7. A reactor module (1) as claimed in one of claims 1 to 6, wherein intermediate walls (12), which form in each case a lower, outer prechamber (13) and a lower, inner prechamber (14) in the feed line (3) and an upper, outer prechamber (15) and an upper, inner prechamber (16) in the discharge line (4), are provided in the feed and discharge lines (3, 4), and wherein the heat-exchange medium is fed to the lower, outer prechamber (13), through a region between the feed line (3) and the discharge line (4) to the upper, inner prechamber (16), through its jacket aperture (6) to the reactor space surrounding the catalyst tubes (2), subsequently through the jacket aperture (5) to the lower, inner prechamber (14), through the region between the feed and discharge lines (3, 4) to the upper, outer prechamber (15), and finally back to the pump(s) through the discharge line (4).
8. A reactor module (1) as claimed in claim 7, which has an outer chamber (17) positioned against the broad reactor side assigned to the catalyst tube-free space (8), with apertures (18, 19) to the reactor space surrounding the catalyst tube bundle (2), and with fixed or adjustable passage apertures (20) for the heat-exchange medium in the outer chamber (17).
9. A reactor module (1) as claimed in one of claims 1 to 8, wherein the catalyst tubes of the catalyst-tube bundle (2) are arranged in mutually offset rows, where the ratio of the tube separation  $s_t$  transverse to the inflow direction by the heat-exchange medium to the tube separation  $s_l$  longitudinal to the inflow direction by the heat-exchange medium is preferably greater than or equal to  $2 \cdot \sqrt{3}$ , particularly preferably equal to  $2 \cdot \sqrt{3}$ .
10. A reactor constructed from two or more reactor modules (1) as claimed in one of claims 7 to 9 lined up in rows against one another on the narrow side surfaces in the direction of the longitudinal axes of the catalyst tubes.
11. The use of a reactor module as claimed in one of claims 1 to 9 or of a reactor as claimed in claim 10 for carrying out oxidation reactions, in particular for the preparation of phthalic anhydride, maleic anhydride, glyoxal, (meth)acrolein or (meth)acrylic acid.

**Abstract**

A reactor module (1) having a catalyst-tube bundle (2) is proposed through whose space surrounding the catalyst tubes a heat-exchange medium circuit is run, with  
5 feed and discharge lines (3, 4) at both ends of the reactor module, where the reactor module (1) has a rectangular cross section. Reactor modules (1) can be lined up in rows against one another in any desired number and thus combined to form reactors of the desired capacity.

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(Figure 1)

O.Z. 0050/49633/WW

A perspective view of a multi-layered rectangular device, likely a battery or capacitor. The device consists of several stacked layers. The top layer (1) is a solid plate. Below it is a layer (2) containing a grid of circular elements (3). This is followed by a layer (4) with a grid of rectangular elements (5). The bottom layer (6) is another solid plate. The device is shown with rounded ends (7) and a central vertical slot (8). Arrows (9) indicate the flow of current or ions through the layers. Labels P, W, and G are used to identify specific components or regions.



FIG.3

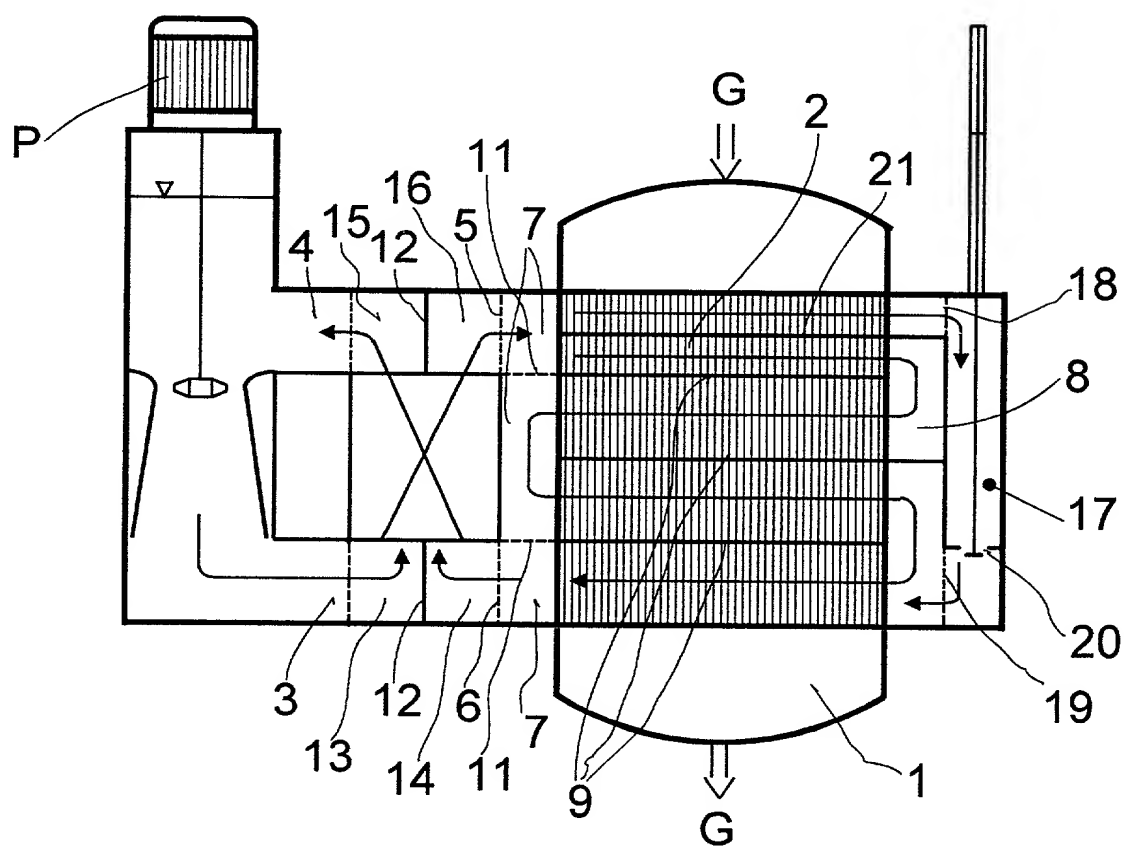






FIG.5

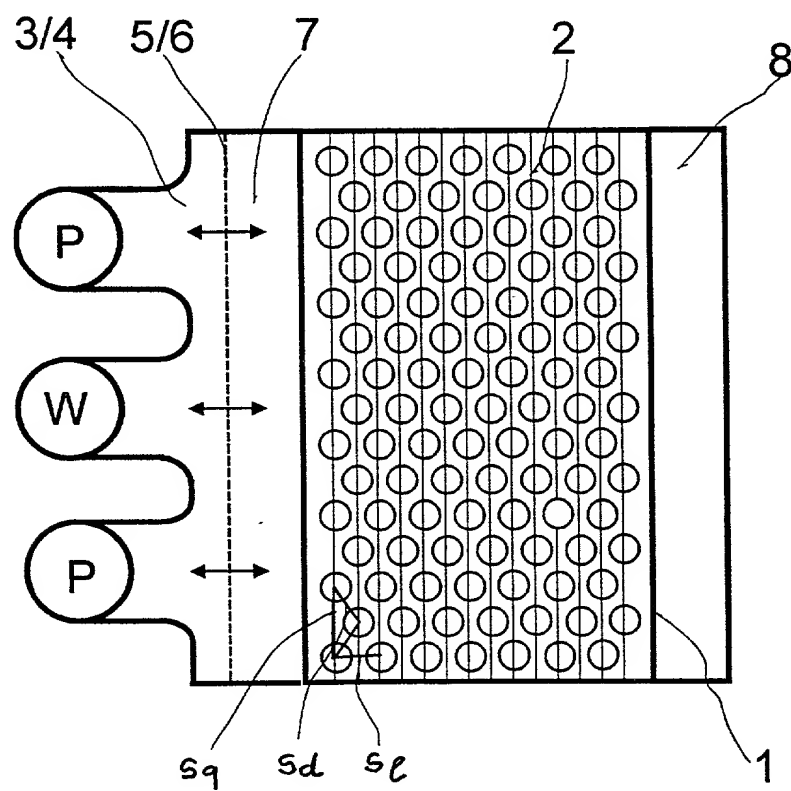
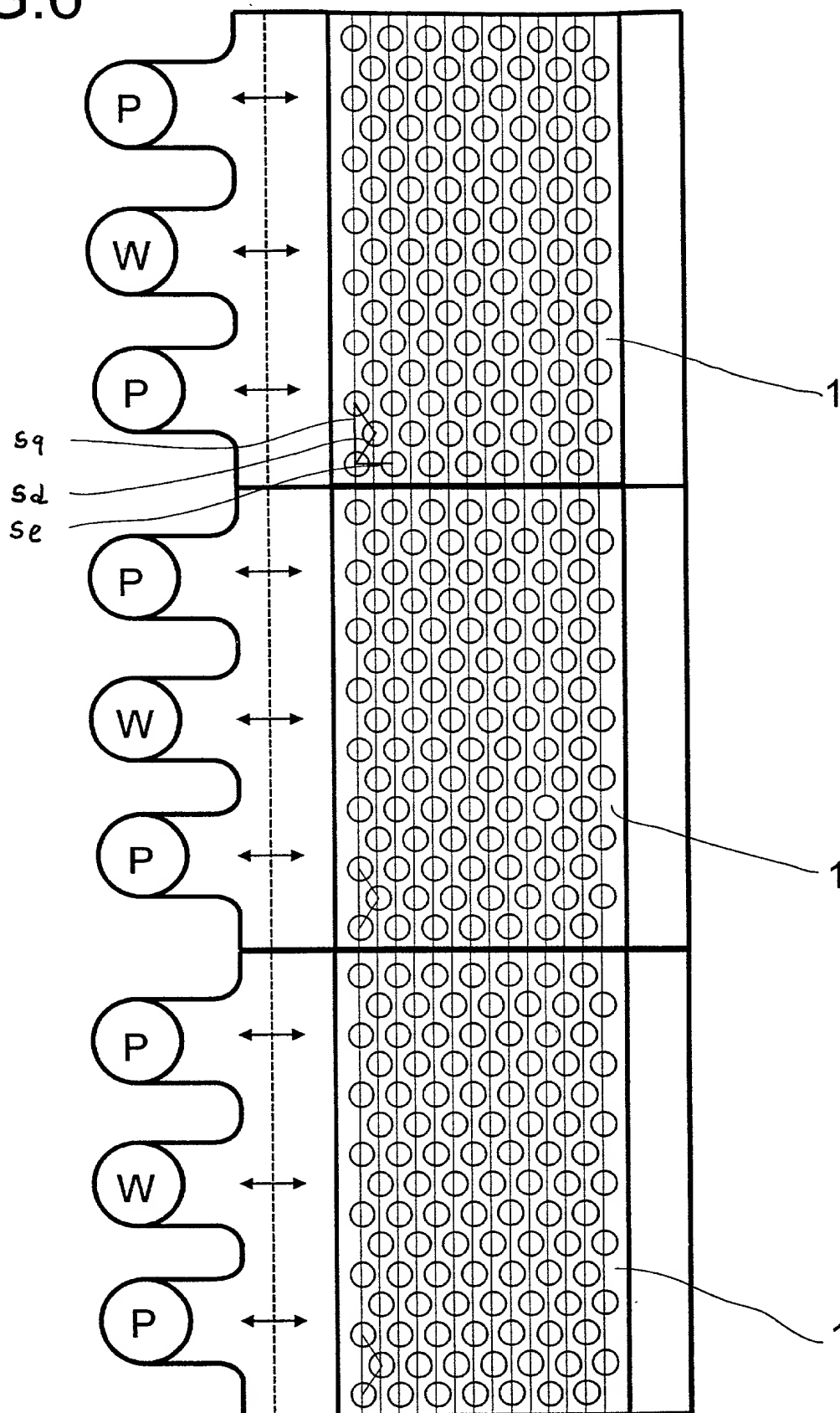


FIG.6



# Declaration, Power of Attorney

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0050/049633

We (I), the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Reactor module having a catalyst-tube bundle

the specification of which

☒ is attached hereto.

☐ was filed on \_\_\_\_\_ as

Application Serial No. \_\_\_\_\_

and amended on \_\_\_\_\_.

☒ was filed as PCT international application

Number PCT/EP99/09971

on 15/12/1999

and was amended under PCT Article 19

on \_\_\_\_\_ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)–(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
19857842.3	Germany	15 December 1998	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No

We (I) hereby claim the benefit under Title 35, United States Codes, § 119(e) of any United States provisional application(s) listed below.

\_\_\_\_\_  
(Application Number)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Application Number)

\_\_\_\_\_  
(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.

Filing Date

Status (pending, patented,  
abandoned)

_____	_____	_____
_____	_____	_____
_____	_____	_____
_____	_____	_____

And we (I) hereby appoint **Messrs. HERBERT. B. KEIL**, Registration Number 18,967; and **RUSSEL E. WEINKAUF**, Registration Number 18,495; the address of both being Messrs. Keil & Weinkauff, 1101 Connecticut Ave., N.W., Washington, D.C. 20036 (telephone 202-659-0100), our attorneys, with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the patent, and to transact all business in the Patent Office connected therewith.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date 14/03/2001

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